Atomistic Studies of Quasi-Static Compression in α-HMX

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We have performed molecular dynamics simulations of uniaxial compression for the α conformer of octahydro-1,3,5,7-tetranitro-1,3,5,7-tetrazocine (α HMX) along different crystallographic directions. The system studied contained 24576 molecules (688128 atoms) in a box with dimensions 180.8 x 192.7 x 194.7 Å. Periodic boundary conditions were employed in all directions. The force field used was taken from Smith and Bharadwaj [1] and includes all degrees of freedom with the exception of C-H bond stretching vibrations, which were fixed at their equilibrium values using the SHAKE algorithm. We performed the simulations using a modified version of the LAMMPS Molecular Dynamics code [2, 3] from Sandia National Laboratories. The system was thermalized for 10 ps at 300 K in the NVT ensemble before starting the compression cycles. Each compression cycle consisted of a uniaxial decrease in volume of 0.1% relative to the original volume V_0 followed by 0.4 ps of NVE equilibration. A total of 250 cycles were performed, producing a final system with a volume that was $0.75V_0$. After compression, the system was expanded to its original volume at the same rate and with the same equilibration as followed during the compression. Compression/expansion cycles were performed for the (100), (010), and (001) directions.

A large degree of directional anisotropy in response to compression was observed. Figure 1 shows pressure vs relative volume for the compression and expansion cycles in all three directions. For compression in the (100) direction, the system begins to exhibit plastic deformation while maintaining the α crystal structure at a pressure of about 2.0 GPa, with planes slipping in the ½(101) direction (Fig. 2a). For compression in the (010) direction a phase transition is observed at a pressure of about 0.5 GPa (not shown), while during compression in the (001) direction a different phase transition is observed at about 1.0 GPa (Fig. 2b) with a further rearrangement at a pressure just below 2.0 GPa. The two phase transitions observed during compression in the (010) and (001) directions persisted even after the system was expanded back to its original volume.

In all cases compression increases the number of first near-neighbors (defined as those with molecular center-of-mass distances less than 9.5 Å) from 14 to 16. Up to 8% of the molecules change ring conformation during compression; most of those that do are localized to the neighborhood of the slip planes, grain boundaries, or "twin" planes [(001, (010), and (001) compressions, respectively].

There are no experimental data to confirm or refute the results reported here, but the robustness of the force field for thermophysical and elastic mechanical properties where data do exist gives us some confidence that the directional anisotropy, plastic deformation and phase transitions are qualitatively representative of what would be observed in the real material during uniaxial quasi-static compression. The results shown here serve a basis for the large-scale molecular dynamics shock compression computations described in another paper by Jaramillo and Sewell in this volume on p. 133.

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[1] G.D. Smith and R.K. Bharadwaj, *J. Phys. Chem. B* **103**, 3570 (1999).
[2] S.J. Plimpton, *J. Comp. Phys.* **117**, 1 (1995).
[3] S.J. Plimpton, et al., *in Proc. of the Eighth SIAM Conference on Parallel Processing for Scientific Computing,* (Minneapolis, MN, March 1997).

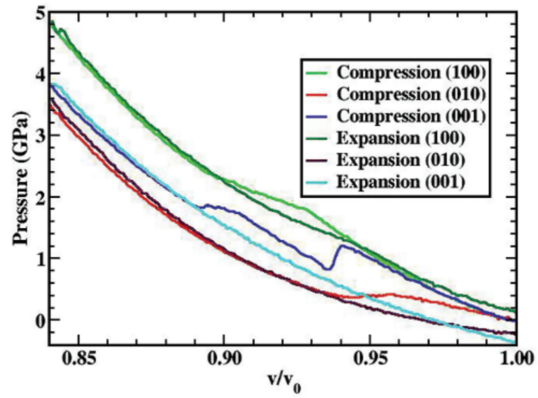

Fig. 1.
Pressure response of α HMX during quasi-static uniaxial compression and expansion.

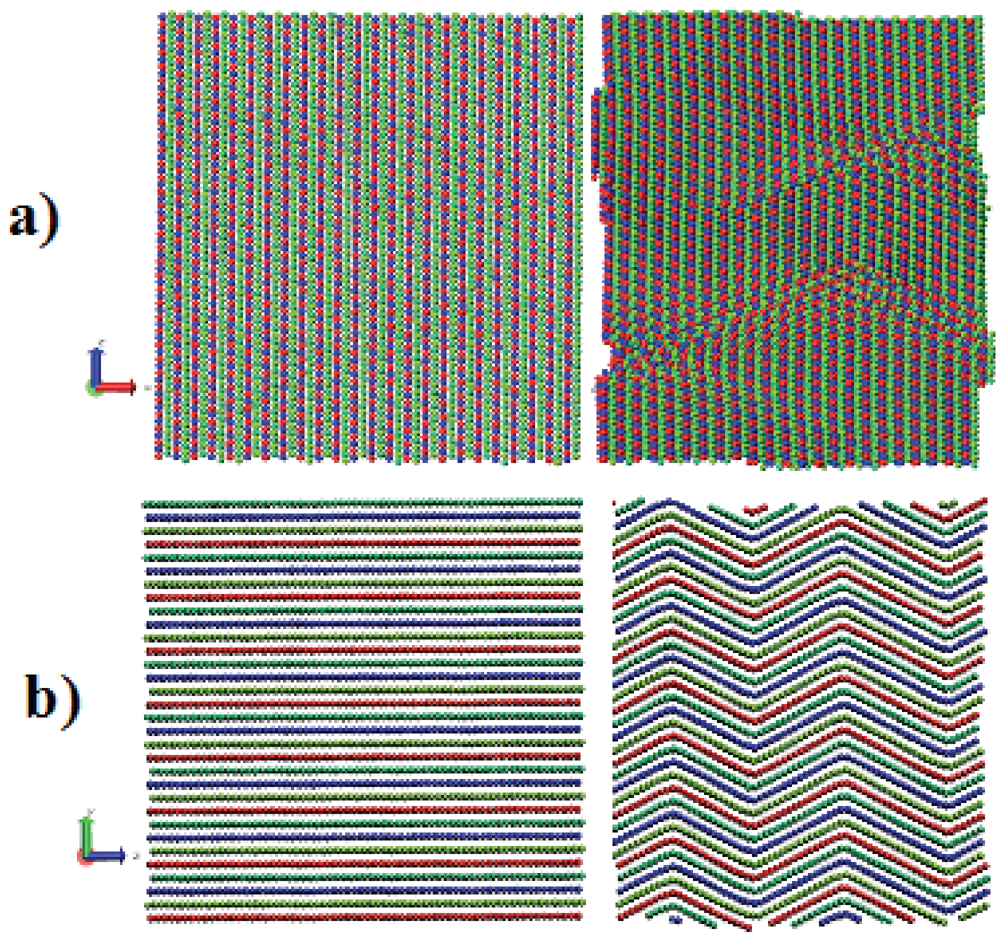

Fig. 2.
Snapshots of the α HMX system before (right) and after (left) compression in the (100) direction (a) and (001) direction (b). For simplicity, the snapshots show only the center of mass of the molecules, although fully atomistic simulations were performed. Molecules were colored only to facilitate the observation of the features created by compression.